

Athermal rheology of weakly attractive soft particles

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We study the rheology of a soft particulate system where the inter-particle interactions are weakly attractive. Using extensive molecular dynamics simulations, we scan across a wide range of packing fractions (ϕ), attraction strengths (u) and imposed shear-rates ($\dot{\gamma}$). In striking contrast to repulsive systems, we find that at small shear-rates generically a fragile isostatic solid is formed even if we go to $\phi \ll \phi_J$. Further, with increasing shear-rates, even at these low ϕ , non-monotonic flow curves occur which lead to the formation of persistent shear-bands in large enough systems. By tuning the damping parameter, we also show that inertia plays an important role in this process. Furthermore, we observe enhanced particle dynamics in the attraction-dominated regime as well as a pronounced anisotropy of velocity and diffusion constant, which we take as precursors to the formation of shear bands. At low enough ϕ , we also observe structural changes via the interplay of low shear-rates and attraction with the formation of micro-clusters and voids. Finally, we characterize the properties of the emergent shear bands and thereby, we find surprisingly small mobility of these bands, leading to prohibitively long time-scales and extensive history effects in ramping experiments.

I. INTRODUCTION

Soft jammed materials (e.g. foams, grains, gels, emulsions, colloids, etc.) are commonplace in nature and our daily lives, exhibiting a wide range of rheological behaviour. Due to large scale industrial applications and also abundance in natural phenomena, understanding the flow properties of these soft materials is an area of intense research. Despite the wide variety of materials, many such systems exhibit the phenomenon of *jamming*, which is a non-equilibrium transition [1, 2] whereby the material becomes solid when the volume fraction, ϕ , of the constituent particles crosses a threshold, i.e. the jamming point ϕ_J . Above ϕ_J , a stress threshold (known as yield stress), needs to be exceeded to obtain a steadily flowing state. In the context of rheology, the development of a finite yield stress constitutes the onset of jamming at $\phi = \phi_J$ [3]. To understand and develop theories for this phenomenon, systems with simplified particle interactions have served as models. In particular, interactions based on repulsive, and frictionless contacts have been studied extensively [1, 2]. It is now well known that in granular systems, the presence of frictional interactions modify the jamming phase diagram [4] and jamming becomes possible in a range of volume fractions depending on the preparation as well as on the inter-particle friction coefficient [5]. The role of attractive interactions in determining the rheology of such complex fluids has, however, only recently commenced [6–10].

In the case of granular materials, attraction can appear in different ways, like the development of capillary bridges [11–13] or van der Waals forces [14, 15], and it might change the rheology of the system significantly. For example, a finite yield stress is observed in some attractive systems at packing fractions much less than the repulsive ϕ_J [9, 16–18]. Furthermore, shear-banding, i.e. the occurrence of spatially inhomogeneous flow [19, 20], is often observed in these materials. While shear local-

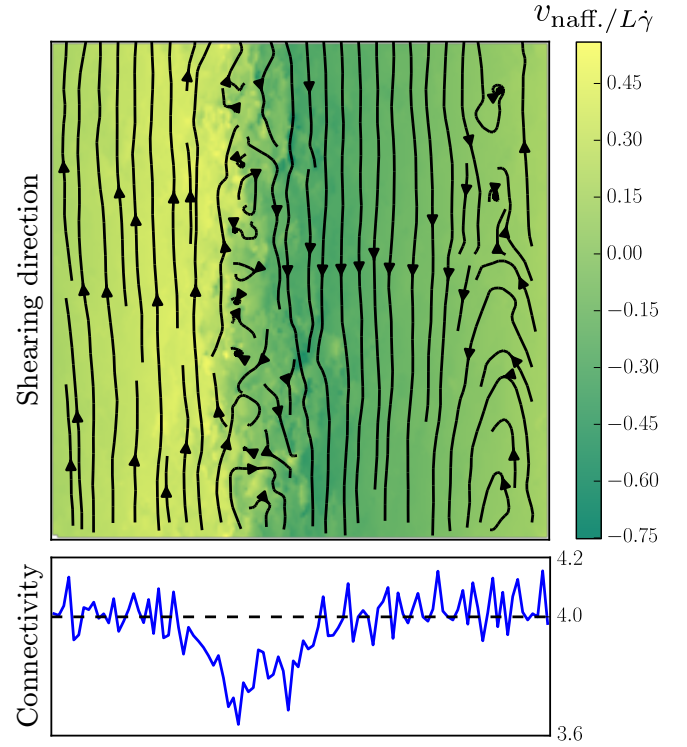


FIG. 1. (Color online, top), A snapshot of the non-affine velocity field in a system exhibiting shear-banding, using $N = 2 \times 10^4$ particles, $\phi = 0.82$, $u = 2 \times 10^{-5}$ and $\dot{\gamma} = 2.5 \times 10^{-6}$. The coloring is based on the velocity in the shearing direction. Stream lines show the non-affine flow field. (Bottom), the corresponding connectivity profile. It reveals that in the solid band, connectivity fluctuates around the isostatic value but decreases in the fluidized band.

ization has been reported in both repulsive and attractive systems [17, 21], it has now been established that dense systems of soft repulsive particles, studied over

sufficiently long times, do not exhibit shear bands as a permanent feature [21–23]. For the occurrence of permanent shear bands, non-monotonic constitutive laws are necessary [19, 24], which is not observed in repulsive systems. On the other hand, in an earlier work [6], we have demonstrated how such non-monotonic flow curves can be obtained by including weak attractive interactions. Therefore studying such systems can shed light on physical mechanisms behind the observation of permanent flow heterogeneities in athermal jammed materials.

In our previous work [6], using numerical simulations, we investigated the effect of weak attractive interactions on the rheology of granular systems. We observed the development of a finite yield stress below the jamming point and also the occurrence of non-monotonic flow curves leading to permanent shear bands (Fig. 1). We also proposed a simple theoretical model to rationalise the observations, based on the competition between shear-induced fluidization and the tendency of aggregation. In the current study, we concentrate on extending the jamming phase diagram, and demarcate the regimes where shear-banding can be observed. We also discuss the associated flow properties, local structure and particle dynamics at microscopic level in greater details, specifically in the crossover from attraction-dominated to repulsion-dominated regime. Further, we investigate the properties of shear bands: the behaviour of the interface as a function of external strain rate, the dynamics of the entire band etc. These help in providing a more coherent picture regarding the occurrence of shear-bands in attractive systems.

The paper is organized as follows. First, we introduce the model and outline the simulation method. This is done in Sect. II. Then, we discuss our results in details in Sect. III. This is carefully structured in order to sequentially discuss our findings starting with analysis concerning macroscopic (system-level) behavior (Sect. III A) and then connecting to microscopic (particle level) aspects, separating out structural (Sect. III B 1) and dynamical (Sect. III B 2) observations. We end our discussions in Sect. III C with a systematic analysis of the formation of flow heterogeneities. At the end (Sect. IV), we conclude and discuss our results.

II. MODEL

We consider a two-dimensional system of N soft disks interacting via the following potential:

$$V(r_{ij}) = \begin{cases} \epsilon \left[\left(1 - \frac{r_{ij}}{d_{ij}}\right)^2 - 2u^2 \right], & \frac{r_{ij}}{d_{ij}} < 1 + u \\ -\epsilon \left[1 + 2u - \frac{r_{ij}}{d_{ij}} \right]^2, & 1 + u < \frac{r_{ij}}{d_{ij}} < 1 + 2u \\ 0, & \frac{r_{ij}}{d_{ij}} > 1 + 2u \end{cases} \quad (1)$$

where r_{ij} is the distance between the i th and j th particles, and $d_{ij} = (d_i + d_j)/2$ is the summation of their

radii. Thus, there exists a harmonic repulsive interaction when the particles overlap, $r_{ij} < d_{ij}$. Additionally, there is a short-range attractive interaction between the particles when the distance is within some threshold, $d_{ij} < r_{ij} < d_{ij}(1 + 2u)$. The parameter u is introduced to characterize the width ($2u$) and also the strength (ϵu^2) of the attractive potential. The scale for attractive forces is then $\epsilon u/d$. Thus, attractive forces are characterized by just a single parameter. This greatly reduces the computational complexity and at the same time keeps the model as simple as possible. The inter-particle potential and the corresponding force is illustrated in Fig. 2, for a choice of the parameters ϵ, u .

By choice, we use a simple interaction model which is suitable as a case-study for the sole effect of switching on the attractive interactions between particles. In the first approximation, such a model would be appropriate for attractive emulsion droplets or sticky grains. In reality, granular materials have much more complicated interactions. For example, grains interact via frictional forces [25–27]. These are not considered in this work. The additional complications arising from frictional interactions, e.g. hysteresis [4] or discontinuous shear thickening [28], are therefore excluded for now. We also note that for emulsions, pastes, colloids etc., the frictionless athermal attractive model provides a good description.

In addition to the conservative force, a dissipative force acts between pairs of particles. This viscous force is proportional to their relative velocity and acts only when particles overlap, i.e. $r_{ij} < d_{ij}$,

$$\vec{F}_{\text{diss.}} = -b[(\vec{v}_i - \vec{v}_j) \cdot \hat{r}_{ij}] \hat{r}_{ij} \quad (2)$$

where b is the damping coefficient. In our simulations, $b = 2$ which indicates that our system is overdamped. We also explore the rheology for other values of b , which we discuss later in the text.

To investigate the rheology of such a system of particles, we perform molecular dynamics simulations using LAMMPS [29].

In order to avoid crystallization, we choose a 50:50 binary mixture of particles having two different sizes, with a relative radii of 1.4. The system is sheared in \hat{x} direction with a strain rate $\dot{\gamma}$ using Lees-Edwards boundary conditions. The volume fraction is $\phi = \sum_{i=1}^N \pi R_i^2 / L^2$, where R_i is the radius of the i -th particle and L is the length of the simulation box. A wide range of volume fractions, from $\phi < 0.50$ to $\phi = 1.0$ has been investigated, and the (repulsive) jamming transition for this system occurs at $\phi_J \sim 0.8430$. Different systems sizes have also been studied, viz. $N = 1000$ and 20000 . Most of the results are reported for $N = 1000$. For the case of analysing the formation of shear-bands, we need to consider a larger system, for which we use $N = 20000$.

The unit of energy is ϵ and the unit of length is the diameter of the smaller particle type, $d = 1.0$. The unit of time is hence $d/\sqrt{\epsilon/m}$, where $m = 1.0$ is the mass of the particles. The velocity-Verlet algorithm is used

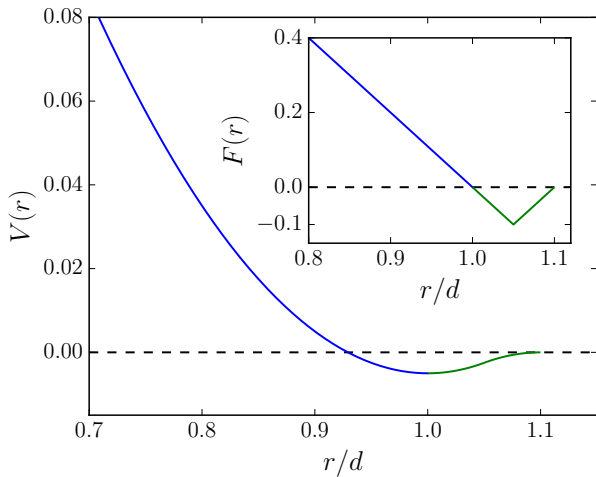


FIG. 2. Interparticle potential $V(r)$, for $\epsilon = 1$ and $u = 0.05$. The inset is the corresponding elastic force. The attractive part is shown in green ($r/d > 1.0$).

to integrate the particles' equations of motion. All measurements are done, after steady flow has been reached. Typically, for large ϕ , measurements are done over strain intervals of 6, after a transient initial strain of 2. In the case of smaller ϕ , measurements are done over a strain window of 10 – 15, after an initial transient strain of 5 – 10. We ensure steady state conditions, wherein the observables typically fluctuate around a constant mean value.

III. RESULTS

To analyse the effect of weak attractive interactions, we study the macroscopic physical properties as well as structure and dynamics of the system at the microscopic level, scanning across a wide range of densities and attraction strengths.

A. Macroscopic Rheology

1. Flow Curves

The macroscopic rheological response of the system of particles is characterised by measuring the stress (σ) that is generated under the application of external shear rate ($\dot{\gamma}$). When a system of repulsive particles is sheared at $\phi < \phi_J$, having the dynamics described, the rheological response shows Bagnold scaling, i.e. $\sigma \sim \dot{\gamma}^2$. In Figure 3, the dashed line shows such a flow curve for the repulsive system (with parameters $u = 0.0$, $\phi = 0.65$ and $N = 1000$). The question that we address is how such a flow curve is affected by introducing attractive interactions between the particles. As shown in our previous work, the material becomes rigid with the appearance of a yield

stress, $\sigma_y = \sigma(\dot{\gamma} \rightarrow 0)$, when attraction is added [6]. This can be seen in Figure 3. When a weak attraction ($u = 2 \times 10^{-4}$) is switched on, a finite σ_y emerges at $\phi = 0.65$. We also observed that with increasing ϕ at this attraction strength, σ_y increases. Thus, for such weakly attractive systems, rigidity sets in at volume fractions much below the repulsive jamming point $\phi_J = 0.8430$ [6, 8]. Above the jamming point, flow curves follow a standard Herschel-Bulkley form which is consistent with earlier results [7].

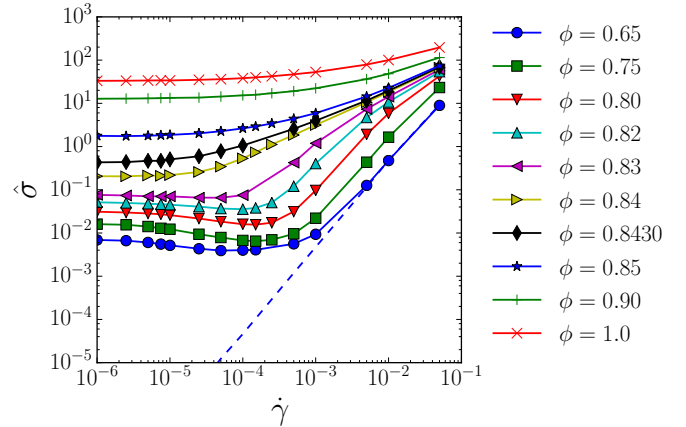


FIG. 3. Flow Curves: normalized shear stress $\hat{\sigma} = \sigma d / \epsilon u$ as a function of strain rate $\dot{\gamma}$ for different volume fractions. The dashed line corresponds to the repulsive system, $u = 0.0$. Solid lines represent attractive systems with the attraction range $u = 2 \times 10^{-4}$. For such systems, the jamming transition occurs around $\phi_J \approx 0.8430$

The influence of the attractive interactions is observed only in the regime of small shear-rates. At large shear rates, the dependence of σ on $\dot{\gamma}$ is identical, for the attractive and for the repulsive systems. One can see this in Figure 3, for $\phi = 0.65$. Thus, for attractive systems, we refer to the high shear rate regime as “repulsion-dominated” and the low shear rate regime as “attraction-dominated”. In our earlier work, we have also noted that the range of shear-rates over which the “attraction-dominated” regime is observed broadens with increasing the attraction range and decreasing the volume fraction.

A key feature of the flow curves, in the regime of weak attraction, is the existence of a non-monotonic shape. We have shown earlier and rationalised how such a behaviour occurs for $\phi < \phi_J$. The non-monotonic dependence in σ vs $\dot{\gamma}$ leads to a mechanical instability, which shows up in the form of localised shear bands [30, 31]. We will discuss this in more details in later sections.

For attractive particles, since a finite yield stress is exhibited even below ϕ_J , the jamming phase diagram needs to be redrawn. In the top panel of Figure 4, we show the necessary modification in the $\sigma - \phi$ plane, for an attraction strength of $u = 2 \times 10^{-4}$. The solid black line marks the yielding threshold for this attractive strength, with varying ϕ . It can be seen that the system has a

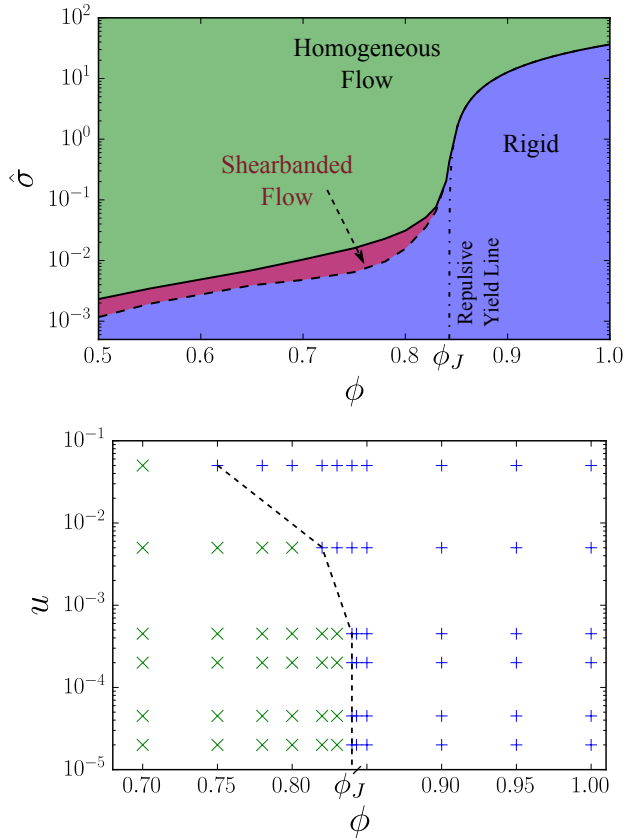


FIG. 4. (Color online, top), The modified jamming phase diagram for attractive systems, choosing $u = 2 \times 10^{-4}$: Solid line marks the yield stress σ_y as a function of ϕ . Above $\phi_J (= 0.8430)$, yield stress behaves as $\sigma_y \propto \delta\phi^\alpha$ where $\alpha = 1.04$. We also demarcate in the phase diagram, the region (shown in purple), where shear-banded flow is expected. (Bottom), The state space of u and ϕ , where points to observe shearbands (cross symbols) or homogeneous (plus symbols) flow are marked. The critical volume fraction of the transition between shearbanded and homogeneous flow decreases with increasing the attraction strength.

finite yield stress even at the smallest ϕ that we have explored, viz. $\phi = 0.50$ (see Figure 4). This is in contrast to the repulsive system where shear rigidity sets in at ϕ_J . As indicated in the figure, for $\phi > \phi_J$, σ_y vs ϕ is identical for (weakly) attractive and repulsive systems. We also mark in the same figure, using dashed lines, the regime in which non-monotonic flow curves are observed for this attraction strength; for each ϕ , this line marks the location of the minimum in the non-monotonic flow curve. Thus, in this regime, shear-banded steady-state flows are expected.

The range of attraction over which flow instabilities are observed, for varying attraction, is indicated in the bottom panel of Figure 4. We note that with decreasing ϕ , the range of attraction strengths over which non-monotonic behaviour is observed starts to increase and for $\phi = 0.7$, such a rheological response occurs for all

attraction strengths that we have explored.

As is depicted in Figure 4 (top), for attractive particles, a finite yield stress exists even far below the jamming point where the yield stress vanishes for repulsive particles. Thus, a valid question is whether there is a threshold in volume fraction above which a rigidity transition, with a finite yield stress, occurs for weakly attractive systems. To answer this question, one needs to go to smaller ϕ than the range depicted in the phase diagram. However, for $\phi < 0.50$, we observe that the system shows strong sensitivity to the initial configuration and defining a steady state to measure the yield stress becomes increasingly difficult. In such systems, particles frequently form a large cluster and flow for a while, or form two or more smaller clusters which do not interact most of the time, due to the large void spaces between them; see Fig. 5. Therefore the stress measurements become very sensitive to the strain window over which the measurement is performed. Therefore, in this work, we focus only on relatively denser systems ($\phi \geq 0.50$) where situations as depicted in Fig. 5 do not occur and a steady state is absolutely well defined.

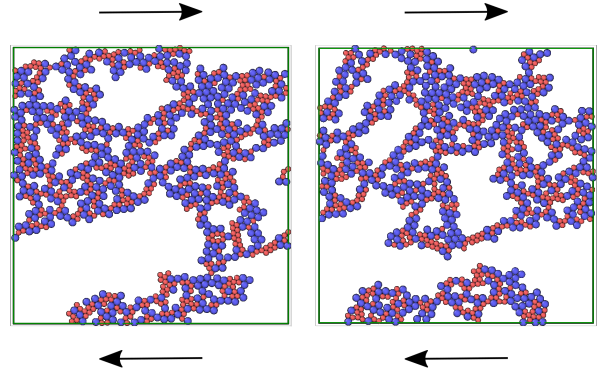


FIG. 5. Two snapshots of a dilute system with $N = 1000$, $u = 2 \times 10^{-4}$ and $\phi = 0.40$. (Left), All particles form a large cluster and flow due to shearing. (Right), Later in the same system, particles form a non-percolating cluster. Arrows indicate the shearing direction.

2. Coordination Number

The flow response is related to the underlying contact network formed by the particles. The structure of the network is characterised by the connectivity, z , which is the average number of contacts per particle. To measure z , we count all neighbors in the range of the interaction potential. Thereafter, we divide these contacts into two different types. When $r_{ij} \leq d_{ij}$, i.e. the particles repel each other, we speak of “repulsive contacts”. With the inclusion of attractive forces, we also define attractive contacts (z_{att}) when $1 < r_{ij}/d_{ij} < 1 + 2u$. The total coordination number is defined as $z = z_{\text{rep}} + z_{\text{att}}$.

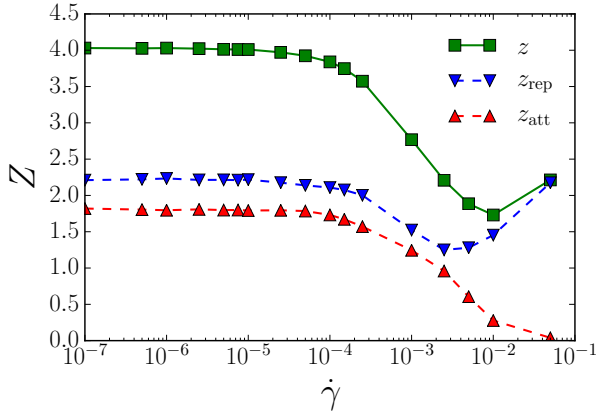


FIG. 6. Variation of different kinds of connectivity, z , z_{rep} , z_{att} , as defined in the text, with strain rate for a system at $\phi = 0.75$ and $u = 2 \times 10^{-4}$. At small strain rates, attractive contacts play important role and keep the total connectivity larger than the isostatic value. At large strain rates, repulsive contacts are dominant, determining the total connectivity behaviour. The drop in $z(\dot{\gamma})$ from isostatic point to the value much smaller, indicates the attractive timescale τ_a .

In Figure 6, for a fixed value of u , we show how z_{rep} , z_{att} and z vary with shear-rate, for an attraction strength where the flow curve is non-monotonic. The data is taken at $\phi = 0.75$, far below ϕ_J . At small strain rates, both z_{rep} and z_{att} are constant, with slightly more repulsive contacts than attractive ones. With increasing strain rate, the attractive contacts rapidly decay and become negligible at large shear-rates. On the other hand, repulsive contacts exhibit a non-monotonic behaviour. While contacts initially get disrupted and thereby decrease with increasing shear-rate, the particles once again get pushed together when the repulsive regime kicks in. Thus, at large shear-rates, z is entirely dominated by the repulsive contacts.

In Figure 7, we plot $z(\dot{\gamma})$ for different ϕ . The dashed line represents the connectivity in a repulsive system at $\phi = 0.65$ and the solid lines corresponds to the attractive systems with $u = 2 \times 10^{-4}$. It is observed that $z(\dot{\gamma}) \rightarrow 0$ at $\dot{\gamma} \rightarrow 0$ for repulsive particles below jamming, which is expected for our model of particles dynamics [32]. On the other hand, for attractive systems and below jamming, as soon as the attraction is introduced, $z_y \equiv z(\dot{\gamma} \rightarrow 0)$ jumps to a value slightly larger than the isostatic connectivity $z_{\text{iso}} = 4$. Consistent with the observation of Khamesh et al [27], no rattlers occur. Figure 7 reveals that such a behavior holds for ϕ even far below ϕ_J . Therefore, finite but small attraction for $\phi < \phi_J$ results in similar isostatic structures as the structure for $\phi = \phi_J$ in repulsive systems. The threshold value $z = 4$ is to be understood as mean-field result and neglects the possibility of mechanisms and states of self stress. There is no reason to believe that these are absent in our networks. However, in line with other studies in the field these effects do seem to play only a minor

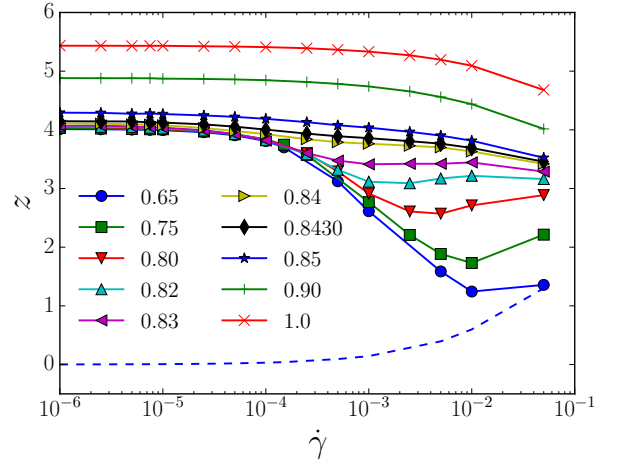


FIG. 7. $z(\dot{\gamma})$ for different volume fractions. Similar to Fig. 3, the dashed line corresponds to the repulsive system $u = 0.0$, solid lines represent attractive system with $u = 2 \times 10^{-4}$.

role [1].

For $\phi > \phi_J$ we find $\delta z \equiv z - z_{\text{iso}} = \zeta_0(\phi - \phi_J)^{1/2}$ with $\zeta_0 \approx 3.78$, consistent with [33]. In the limit of large strain rates, $z(\dot{\gamma})$ reproduces the repulsive results as it is shown for the system at $\phi = 0.65$.

As one can see in Figures 7 and 6, in the limit of small strain rates, $z(\dot{\gamma}) > z_{\text{iso}}$. It remains almost constant with $\dot{\gamma}$ until at a special strain rate, it drops to values far below z_{iso} . This behaviour of connectivity indicates that at small strain rates, attractive forces keep the nearly-isostatic structure of the system stable until the point where the rate of deformation is fast enough to destroy the structure. This rate gives us an attractive time-scale τ_a which is found to scale with the attraction range as $\tau_a \sim 1/u$ independent of ϕ [6].

Recognition of such a timescale also helps in rationalising the variation of stress with shear-rate, as shown in Figure 3. At small $\dot{\gamma}$, the relaxation time is much smaller than the shearing time scale which leads to a continuous reconstruction of the network structure. On the other hand at large $\dot{\gamma}$ this local structure breaks down due to fast shearing and a relatively large relaxation time. The competition between these two mechanisms, attraction induced aggregation and shear induced rupture of local structure, explains the decrease of stress in the intermediate regime.

Now we focus on how the yield stress is related to the connectivity of the system below the jamming point. In elastic spring networks and systems of soft repulsive particles it is well known [1] that the linear response to a macroscopic shear strain γ close to the isostatic point is characterized by strong non-affine motion, expressed by relative particle displacements $\delta_{\perp} \sim \gamma/\delta z^{1/2}$. Those displacements are directed tangentially to the particle contact (see Fig. 8). The corresponding shear modulus of such a linear-elastic response is $g_{\text{lin}} \sim \delta z$. In our system when the contact is not broken, particles see each

other through the harmonic force with a range determined by u . Thus for motion amplitudes smaller than this range, our system can be considered as a network of elastic springs and yielding can be defined as the point where the motion amplitude is larger than the range of attraction and breaks the contact.

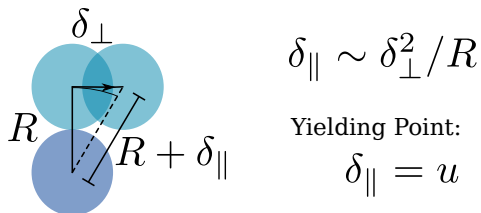


FIG. 8. Sketch of a pair of particles at the yielding point.

To determine the yielding point, we need non-linear loading conditions where the particle's tangential motion is associated with higher-order longitudinal contributions, $\delta_{\parallel} \sim \delta_{\perp}^2/R$ (Pythagoras) (similar reasoning has been applied in different contexts, see [34, 35]). In our attractive system, the maximum dilational strain should be smaller than the attraction range, $\delta_{\parallel} < u$ (Figure 8). The yield strain can then be expressed as $\gamma_y \sim (u\delta z)^{1/2}$. Since the yield stress is $\sigma_y \sim g_{\text{lin}}\gamma_y$, one can write the following scaling relation for the yield stress, attraction range and the distance to the isostatic point:

$$\sigma_y \sim u^{1/2}\delta z^{3/2} \quad (3)$$

The weak attractive forces we use in our model result in the formation of a fragile solid (nearly-isostatic network of particles). The mechanical response of this fragile solid is explained by Equation 3. Figure 9 shows that Equation 3 holds nicely for systems with a wide range of volume fractions below and above the jamming point and different attraction ranges (Dashed line). Above the jamming point and at high enough u , the well-known repulsive behaviour $\sigma_y \sim |\delta\phi|^\alpha$ is observed. It can be understood by noting the fact that in highly dense systems, the repulsive term of Equation 1 is dominant.

3. Potential Energy

The attractive interactions introduce a new energy scale, ϵu^2 . We now investigate how this shows up in the rheology. In the top panel of Figure 10, we plot how the potential energy per particle, E , varies with imposed shear-rate, for various ϕ , using $u = 2 \times 10^{-4}$. For $\phi > \phi_J$, the potential energy per particle is dominated by repulsive contributions and is thus positive. For $\phi < \phi_J$, at small $\dot{\gamma}$, E becomes negative as attractive forces become responsible for stabilising the solid. However, with

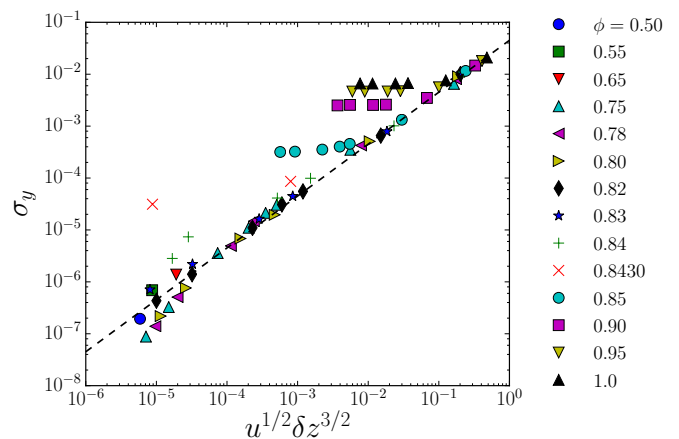


FIG. 9. Yield stress σ_y as a function of the combination $u^{1/2}\delta z^{3/2}$. For small ϕ or u , Eq. 3 holds (dashed line). For large ϕ or u , the yield stress shows the expected repulsive behaviour $\sigma_y \sim |\delta\phi|^\alpha$.

increasing $\dot{\gamma}$, again the particles come into physical contact and the energy becomes positive. In fact, one can see that at $\phi = 0.65$, for large $\dot{\gamma}$, the potential energy per particle for these attractive particles is the same as for the repulsive particles (shown with dashed lines). Since the average number of contacts is z , the variation of the potential energy per particle due to the attractive bonds with changing shear-rate can be written as

$$E = -\epsilon u^2 z(\dot{\gamma}). \quad (4)$$

Earlier, in Figure 6, we had seen that $z(\dot{\gamma})$ is almost constant in the small $\dot{\gamma}$ regime, below $\phi < \phi_J$. Furthermore, it also does not vary much with ϕ (see Figure 7). Thus, in this regime, the potential energy per particle is also expected to be constant, which is illustrated in the bottom panel of Figure 10 for different ϕ .

Similar to defining a stress threshold for yielding to occur, one can define a yield potential energy as $E_y = E(\dot{\gamma} \rightarrow 0)$. The variation of the estimated E_y with ϕ for different attraction strengths is shown in Figure 11. For $\phi > \phi_J$, where repulsion dominates, we see that $E_y \propto (\phi - \phi_J)^\beta$ where we find $\beta \approx 2.1$. For repulsive particles, $E \propto \sigma^2$, implying that $\beta = 2\alpha$ which is consistent with our observations. For $\phi < \phi_J$, from Equation 4, one expects $|E_y| \sim u^2$, and this is demonstrated in Figure 11(c).

4. Shear Stress Ratio

The isostatic solid formed due to attractive interactions, at $\phi < \phi_J$, is resistant to plastic deformations under shear. In granular mechanics, this macroscopic resistance (or friction) is quantified via the ratio of shear-

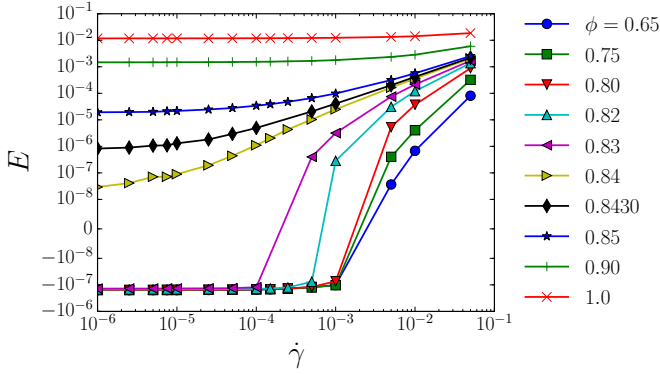


FIG. 10. Potential energy per particle E as function of strain rate $\dot{\gamma}$ for different volume fractions. The dashed line represents the repulsive system with vanishing energy in the limit of zero strain rate. Solid lines are associated with attractive systems with the attraction range $u = 2 \times 10^{-4}$.

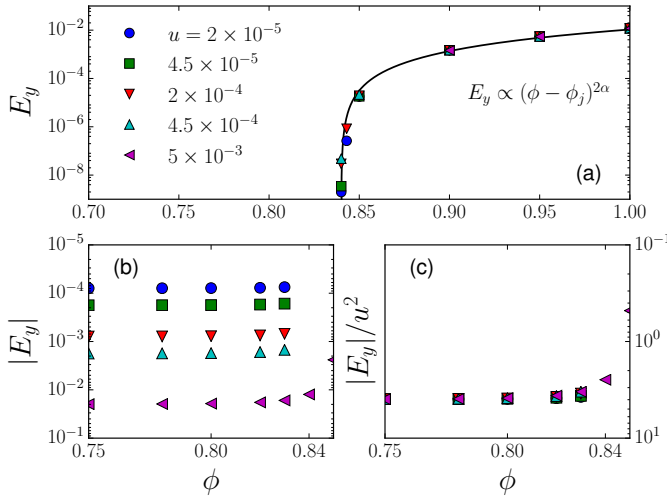


FIG. 11. Yield potential energy E_y as a function of ϕ for different attractions u in (a) repulsion-dominated regime, $\phi > \phi_J$ and (b) attraction-dominated regime, $\phi < \phi_J$. (c) Scaled $E_y(\phi)$ by u^2 in the attraction-dominated regime.

stress to pressure:

$$\mu = \frac{\sigma_{xy}}{P}. \quad (5)$$

To aid our discussion on the macroscopic resistance, we also show the data for pressure, using a rescaled form, in Figure 12, for the same set of ϕ for which shear-stress is shown in Figure 3. The non-monotonic shape of the pressure curves look similar to that of shear stress, the quantitative comparison of which is captured by μ , discussed below. The striking observation is that at $\phi = 0.65$ the pressure curve becomes discontinuous on the log-scale. In that window the pressure is actually negative, implying the dominance of internal tensile forces.

Now, we check how the macroscopic friction μ varies

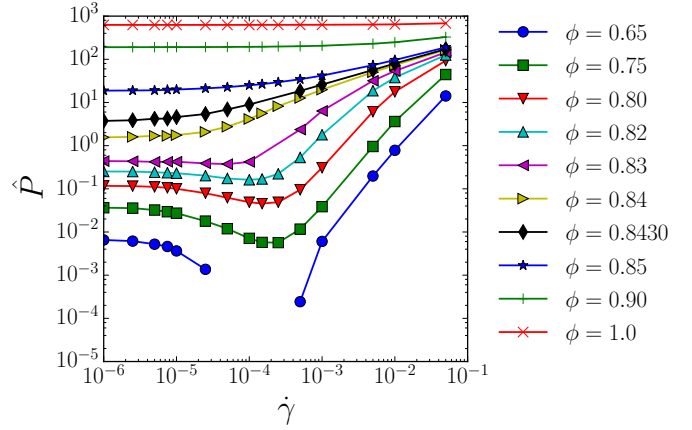


FIG. 12. Rescaled pressure $\hat{P} = Pd/\epsilon u$ as a function of strain rate $\dot{\gamma}$ for different volume fractions. The attraction range is $u = 2 \times 10^{-4}$. The discontinuity in $P(\dot{\gamma})$ at $\phi = 0.65$ corresponds to negative values of P (compressive pressure).

with strain rate scaled by the attractive time-scale, $\dot{\gamma}\tau_a$. In the left panel of Fig. 13 we show the variation of μ for a fixed volume fraction ($\phi = 0.75$) and different attraction strengths. The right panel in Figure 13 shows μ for a fixed attraction ($u = 2 \times 10^{-4}$) but different volume fractions. Both panels highlight the fact that in the presence of finite attraction and for $\phi < \phi_J$, μ develops a peak at the strain rate close to $\dot{\gamma}^*$, where the minimum occurs in flow curves, $\sigma^{\min} = \sigma(\dot{\gamma}^*)$. In fact, due to the rescaling of $\dot{\gamma}$ by τ_a the location of the peak is at the same $\dot{\gamma}^*$ for different u , with the peak height decreasing with increasing attraction, which is in agreement with earlier studies [8, 36]. In the right panel, for $\phi < \phi_J$, this non-monotonic behaviour is observed to disappear for $\phi > \phi_J$. Recalling the discussion above, the apparent discontinuity of μ for $\phi = 0.65$ is due to the pressure becoming negative. The observed non-monotonic behaviour of μ for $\phi < \phi_J$ happens because, under shear, the pressure of the particle assembly drops faster than the shear stress. This also leads to changes in microstructure, which we discuss in a later section.

Also in order to locate our analysis with other works on cohesive grains [10, 27, 37], we have computed the inertial number $I = \dot{\gamma}\sqrt{m/P}$, where m is the mass of the particles and P is the pressure. We find that non-monotonic flowcurves occur for small values of $I < 0.05$, in agreement with [27]. Also the values of the rescaled pressure $\hat{P} = O(1)$, where non-monotonic effects first occur (see Fig. 12), agree with the previous findings (Ref. [27], using longer range attractive interactions, finds shear bands for $\hat{P} < 0.1$).

5. Role of damping

Our main focus is modelling soft athermal materials with attractive inter-particle interactions, like in emul-

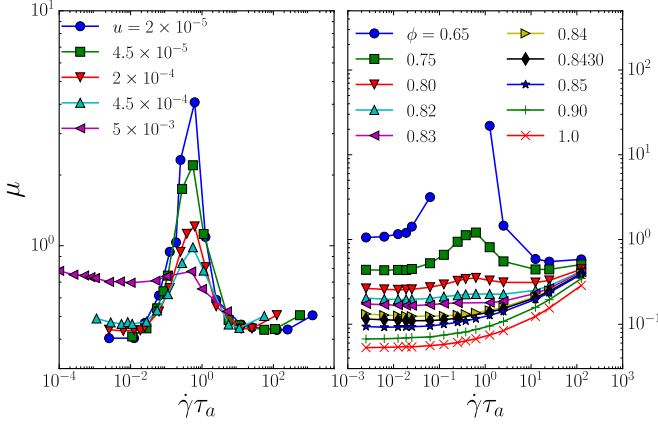


FIG. 13. Shear-stress ratio as a function of $\dot{\gamma}$ for different attraction ranges at $\phi = 0.75$ (left panel) and for different volume fractions at $u = 2 \times 10^{-4}$. Note that for $\phi < \phi_J$, the peak appears close to $\dot{\gamma}^*$ where $\sigma(\dot{\gamma}^*)$ is minimum.

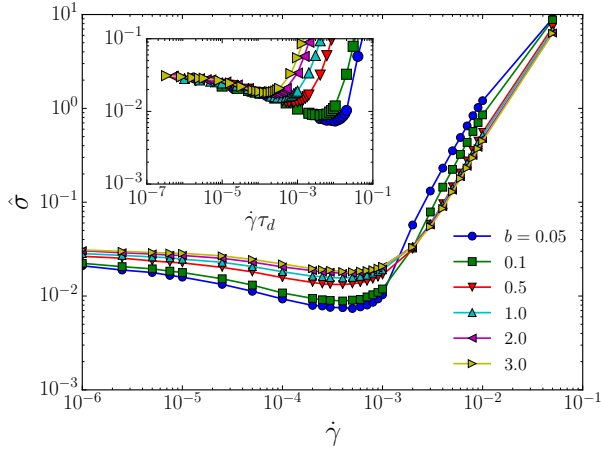


FIG. 14. (Main) Flow curves ($\hat{\sigma} = \sigma d / \epsilon u$), for $\phi = 0.75$ and $u = 7 \times 10^{-4}$, with changing damping coefficient b . (Inset) The low shear-rate regime of the flow curves for different u can be collapsed by using the rescaled variable $\dot{\gamma} \tau_d$, where $\tau_d = m/b$.

sions or gels. In such cases, the motion of the constituent particles is over damped. Thus, in this manuscript, we have reported results for such damped systems, with a relatively large damping parameter $b = 2$. Recent studies have explored how the rheological response changes as one tunes the dynamics from being over-damped, as is the usual case for suspensions, to being under-damped [38, 39]. We now explore this scenario by tuning the damping parameter b . The results are seen in Fig. 14.

We observe that with decreasing the damping coefficient the non-monotonicity in the flow curves become more pronounced. Thus, underdamping enhances the mechanical instability in the system. In effect, by decreasing the damping coefficient, the timescale for energy dissipation increases. This leads to the system being in

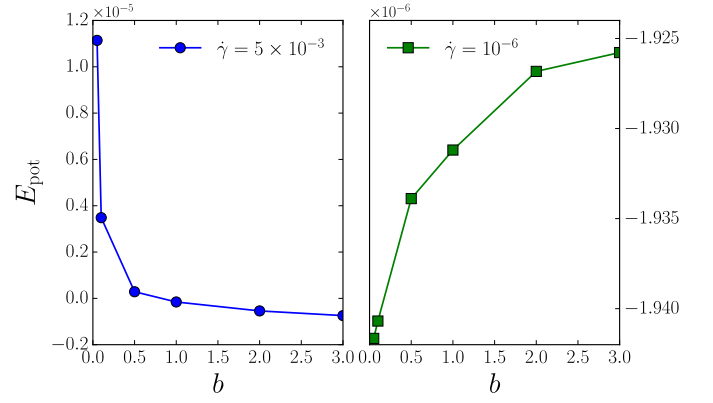


FIG. 15. For $\phi = 0.75$, $u = 7 \times 10^{-4}$, variation of the potential energy per particle E with damping factor b , (left) in the repulsion-dominated regime with $\dot{\gamma} = 5 \times 10^{-3}$ and (right) in the attraction-dominated regime with $\dot{\gamma} = 10^{-6}$.

a fluidized state up to higher strain rates. This can be seen if we rescale the imposed shear-rate $\dot{\gamma}$ with the dissipation timescale defined as $\tau_d = m/b$ (inset of Fig. 14). Thereby, the data for different b can be collapsed in the low shear-rate regime, with the tuning of b leading to the exploration of different regimes along this branch. We have also observed that if we look at the variation of E with changing b , there is a change with decreasing $\dot{\gamma}$. Typically, at large $\dot{\gamma}$, the potential energy per particle of the system decreases as the system gets more damped (i.e. increase of b); quicker dissipation leads to the system not being able to explore all possible higher energy states. However, for $\dot{\gamma}$ in the "attractive regime", we see that E decreases with decreasing b ; the underdamping allowing the system to explore lower energy states in the landscape (Fig. 15). Such a scenario has also been proposed in Ref.[39] to understand how damping influences steady state rheology of amorphous systems and our observations are consistent with that.

B. Structure and dynamics

Next, in order to understand the properties of the system in the different flow regimes, we study the structure and arrangement of particles as well as their dynamics.

1. Structure Factor

Signs of local structures in the attraction-dominated regime can be observed in the structure factor at different $\dot{\gamma}$. The Structure factor $S(q)$ is defined as: $S(q) = N^{-1} \langle \rho(q) \rho(-q) \rangle$, where $\rho(q) = \sum_i^N \exp(i\mathbf{q} \cdot \mathbf{r}_i)$ is the Fourier transform of the number density, N being the total number of particles.

In Figure 16, we plot $S(q)$ at $\phi = 0.75$, $u = 2 \times 10^{-4}$ for different $\dot{\gamma}$. One can observe a small peak at around

$q = 1$. Such a feature corresponds to the formation of clusters of particles, induced by shear. Also, we note that this peak disappears at large shear-rate, implying that this is due to structures formed by the interplay of small shear and attractive interactions.

With the peak appearing at a certain wavenumber q^* , we can define a peak height via $S^* = S(q^*)$. In Figure 17, we show how S^* -data for a range of attraction strengths collapse, if the shear-rate is rescaled with τ_a . This clearly demonstrates the distinct effect that attractive forces have in determining the microstructure over large lengthscales and re-emphasizes the role of the attractive time-scale τ_a .

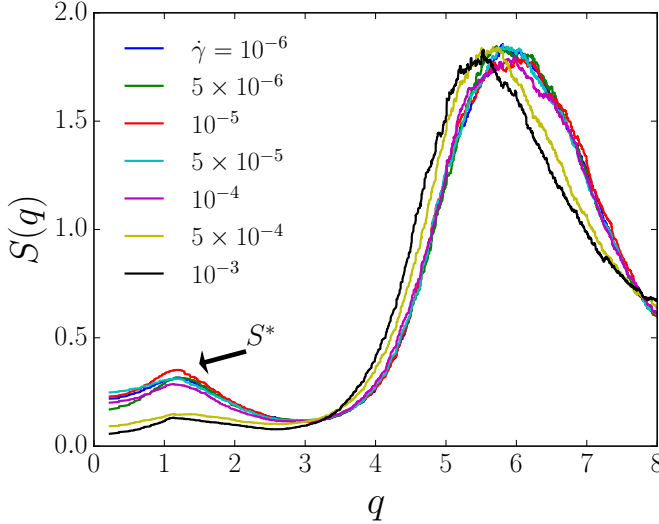


FIG. 16. Structure factor $S(q)$ for different $\dot{\gamma}$ at $u = 2 \times 10^{-4}$ and $\phi = 0.75$. There is a small peak S^* for $q < 2$ associated with the local structure and particle clusters in the attraction-dominated regime ($\dot{\gamma} \leq 10^{-4}$). Since data points are noisy because of the small system size, they have been smoothed using the Savitzky-Golay filter.

For a binary mixture, one can further look at the partial structure factors, $S^{\gamma\nu}(q) = \langle \rho^\gamma(q) \rho^\nu(-q) \rangle$, with $\rho^\nu(q) = \sum_i^{N^\nu} \exp(i\mathbf{q} \cdot \mathbf{r}_i)$ corresponding to the Fourier transforms of the partial density fields. $N^\nu = N/2$ is the number of the sub-population belonging to labels $\nu = 0, 1$, corresponding to small and large particles, respectively. In terms of the partial structure factors, the total structure factor can be written as:

$$S(q) = \frac{1}{2}[S^{00}(q) + S^{11}(q)] + S^{01}(q) \quad (6)$$

The partial structure factors are shown in Figure 18, which shows that the low- q peak in $S_{00}(q)$ is more pronounced than in S_{11} . This implies that smaller particles contribute stronger to the low q peak in $S(q)$, i.e. the mesoscale clustering under shear is caused by the spatial organisation of these small particles (see Fig. 19). It is tempting to attribute this effect to some sort of

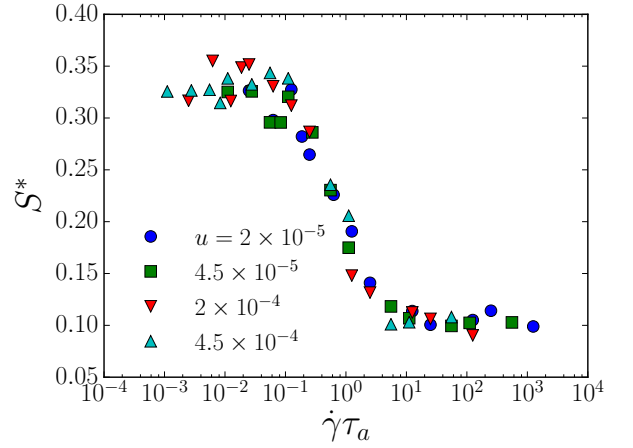


FIG. 17. The first maximum in the structure factor, S^* as a function of $\dot{\gamma}\tau_a$ for a system at $\phi = 0.75$ and different attraction strength. The attractive timescale τ_a is used to rescale the strain rate $\dot{\gamma}$.

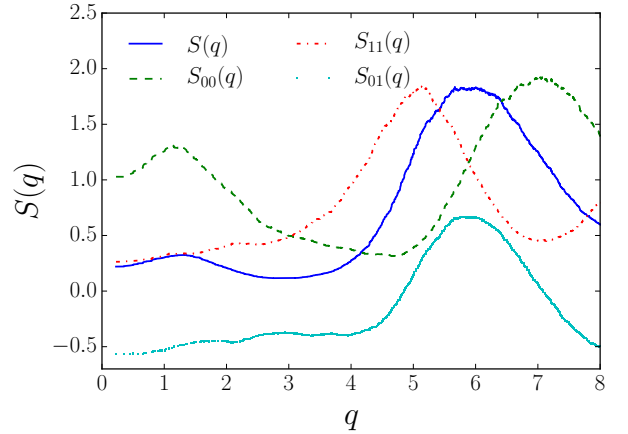


FIG. 18. Partial structure factor $S(q)$ for a system at $u = 2 \times 10^{-4}$, $\phi = 0.75$ and $\dot{\gamma} = 10^{-6}$. S_{00} corresponds to the smaller particles, while $S_{11}(q)$ measures the structure factor of the larger particles.

phase separation under shear. However, we did not observe any substantial growth of these meso-clusters on the time-scales accessible to our simulations.

2. Microscopic dynamics

Similar to the structure of the particles, their dynamics is also affected by introducing attraction. To study this, we investigate the particles' non-affine displacements, which correct for the convective (affine) part of the particle motion, which is induced by the average flow field. At any time t , the non-affine position of a particle

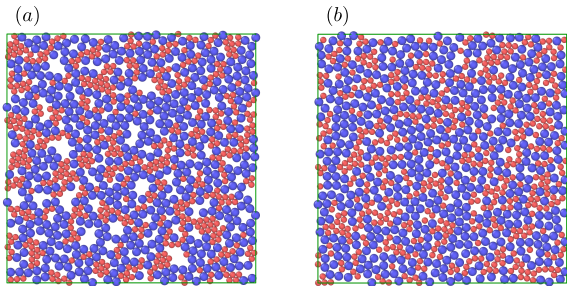


FIG. 19. Snapshots of the sheared system for two different strainrates ((a) $\dot{\gamma} = 10^{-6}$ and (b) $\dot{\gamma} = 10^{-3}$) illustrating the low- q peak observed in the structure factor. For the smaller strainrate, in the attraction-dominated regime, the small particles cluster and the density is rather inhomogeneous, as compared to the larger strainrate, in the repulsion-dominated regime.

can be written as:

$$\mathbf{r}_{\text{naff.}}(t) = \mathbf{r}(t) - \hat{\mathbf{x}} \int_0^t y(t') \dot{\gamma} dt' \quad (7)$$

where $\mathbf{r}(t) = x(t)\hat{\mathbf{x}} + y(t)\hat{\mathbf{y}}$ is the position of the particle at time t . The second part corresponds to the convective contribution, where $\hat{\mathbf{x}}$ is the unit vector in shearing direction and $\hat{\mathbf{y}}$ is the unit vector in gradient direction. Using these non-affine positions, we compute the mean squared displacement (MSD) of the particles, resolved in $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ directions. In Figure 20, we show the corresponding data for $u = 2 \times 10^{-5}$, $\phi = 0.75$.

At large shear rates (Fig. 20(d)), where repulsion dominates, there is no difference in MSD between the shearing and the gradient direction. At large strains, the particles diffuse isotropically. Similarly, near yielding where attraction dominates, the dynamics also seems to be not dependent on direction - in this case, diffusive motion sets in at very large strains, which are somewhat inaccessible to our simulations [Fig. 20(a)]. In contrast, in the intermediate regime, where the flow curve is non-monotonic, we observe that the long-time MSD is different in the flow and the transverse (gradient) directions, i.e. an anisotropy develops. There is an enhancement of non-affine motions in the flow direction, which we relate to the nearby instability towards the formation of shearbands.

To further investigate the nature of the dynamics under shear, we also measured the mean squared non-affine velocity (v^2) in both directions as functions of strain rate. The corresponding data is shown in the top panel of Figure 21. In order to compare the mean squared non-affine velocity of systems at different strain rates, we consider the rescaled quantity $(v/\dot{\gamma})^2$. We observe that this quantity, which measures the extent of the particles' non-affine motion over very short time, increases rapidly with decreasing shear-rate. We also measure the diffusion coefficient (D_y) in the gradient direction, which is shown in

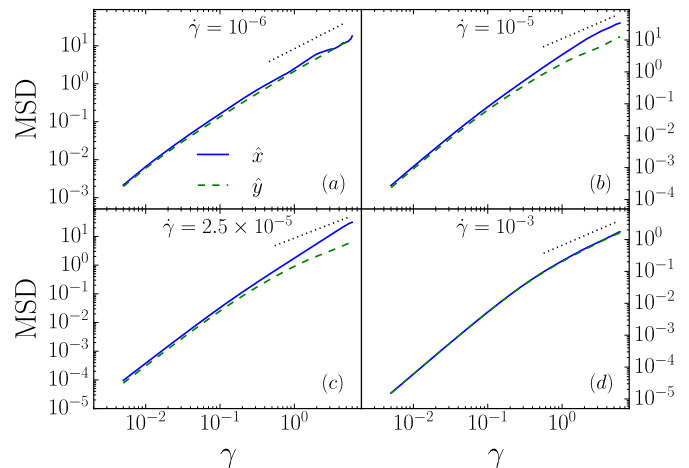


FIG. 20. Non-affine mean-squared displacement (MSD) for a system (a) close to the yield stress, (b) and (c) around the minimum in the flow curve where the attractive and repulsive branches meet and (d) in the repulsion-dominated regime, ($u = 2 \times 10^{-5}$ and $\phi = 0.75$). The dotted line indicates diffusive behaviour.

the bottom panel of Figure 21. This quantifies the extent of non-affine motion over long time and similar to $(v/\dot{\gamma})^2$, has a higher value at smaller shear-rates. The large scatter in the data reflects the approximate nature of calculating the diffusion constant in a regime, where real diffusion is hardly reached. Thus, these observables demonstrate that in the attraction dominated regime, non-affine motion dominates over affine motion, implying that the latter is more energetically costly.

C. Characterizing shear bands

The non-monotonic shape of $\sigma(\dot{\gamma})$ is a signature of a mechanical instability [19, 20, 24, 31]. However, no shear bands are observed in a system of size $N = 1000$ for which the flow curves were presented in Figure 3. It is known that for shear localization to occur, the wavelengths of the unstable modes should be smaller than the system size [30]. The localization of flow into a shear band is demonstrated in Figure 1, which illustrates the velocity field (and corresponding connectivity profile) for a system of size $N = 2 \times 10^4$ at suitable state parameters, viz. $u = 2 \times 10^{-5}$, $\phi = 0.82$ and $\dot{\gamma} = 2.5 \times 10^{-6}$.

We therefore compare how the flow response compares across different system sizes. In Figure 22, we show $\sigma(\dot{\gamma})$ for two different system sizes, viz. $N = 1000, 20000$. For $N = 20000$, we start shearing the system with a random initial configuration at $\dot{\gamma} = 10^{-4}$, ramp it down until $\dot{\gamma} = 10^{-6}$ (data shown in circles) and then again ramp it up (data shown in squares). At each $\dot{\gamma}$, the system is sheared for $\Delta\gamma = 20$ (except at $\dot{\gamma} = 10^{-6}$, where we choose $\Delta\gamma = 12$). Such large strain windows at each $\dot{\gamma}$ try to ensure that we obtain a steady state response at each

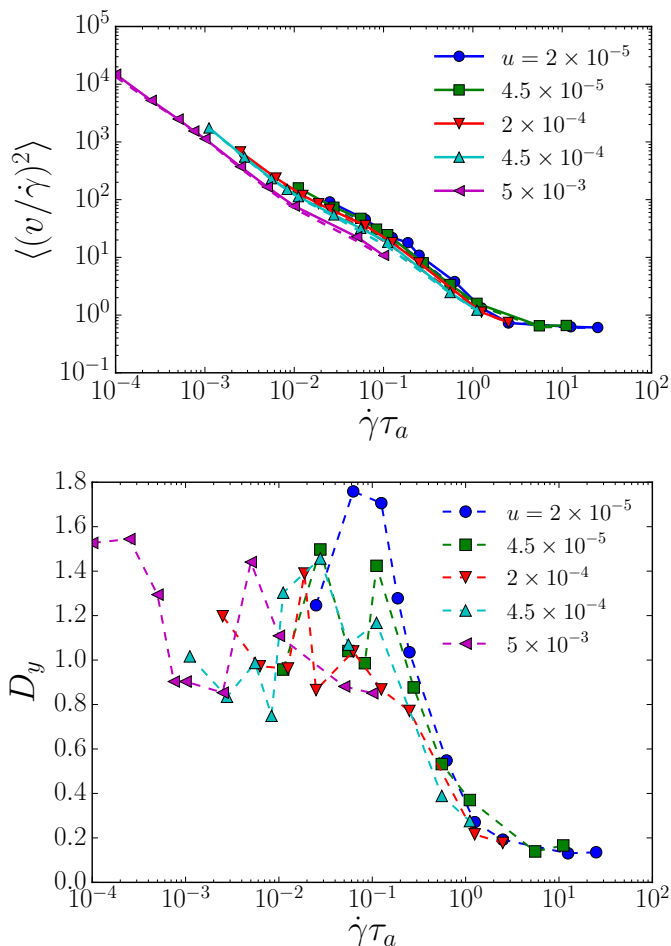


FIG. 21. (Top) Variation of non-affine mean-squared velocity with strain rate (scaled by the attractive time-scale) in shearing and gradient directions for a system at $\phi = 0.75$, for different attraction strengths u . While solid lines represent the shearing direction, dashed lines correspond to the gradient direction. (Bottom) The diffusion constant, measured in gradient direction, as a function of scaled shear-rate, for different u .

state point. The obtained $\sigma(\dot{\gamma})$ data, for the ramp-down and ramp-up, is shown in Figure 22. We observe hysteretic effects similar to findings in recent experiments using the same protocol [40]. Further, we mark with filled symbols the state points at which shear-banding is observed. In comparison to the data for $N = 1000$, the flow curves deviate in the small $\dot{\gamma}$ regime. When the applied shear-rate is ramped down, the non-monotonicity is more pronounced in the regime of small $\dot{\gamma}$. And, when the applied shear-rate is ramped up, the non-monotonicity is nearly suppressed. Further, shear-localization is also visible over a larger range of shear-rates during the ramp-up regime; one needs to go to larger shear-rates to fluidize the solid. It has been suggested that, in the thermodynamic limit, the stress-decreasing part of the flow curve will be replaced by a straight line [20, 30]; our observations are consistent with that trend.

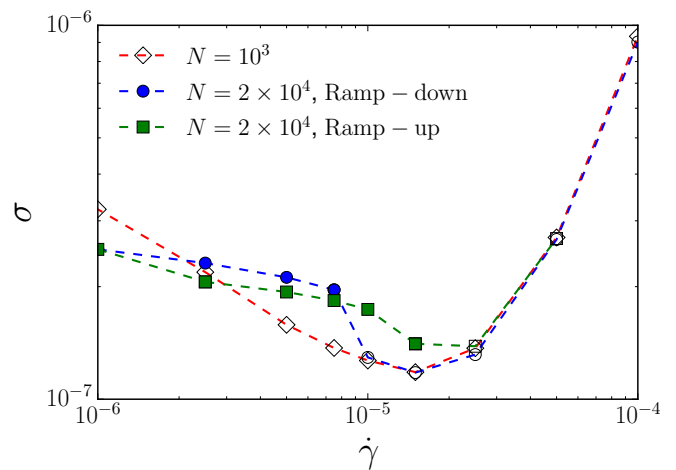


FIG. 22. Flow curves ($\sigma = \sigma d/\epsilon u$) for different system sizes N , at $\phi = 0.82$ and $u = 2 \times 10^{-5}$. The non-monotonic part of the flow curve gets smaller as system size increases and the system is sheared longer. Filled symbols indicate a shear banded flow, open symbols correspond to homogeneous flow.

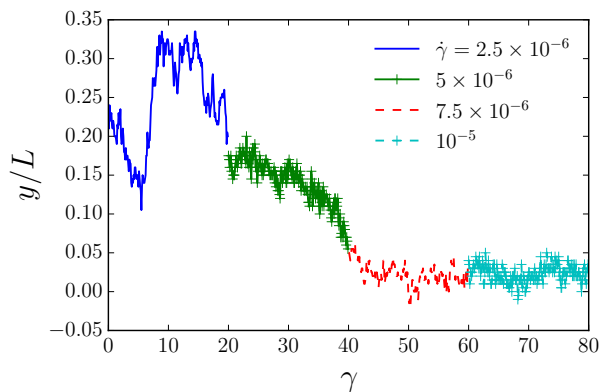


FIG. 23. Position of the center of shear band in time for ramp-up simulations.

We also observe how the spatial location of the fluidized band, in the direction transverse to the applied shear, changes as a function of time, by monitoring the position of the centre of the band. In Figure 23, we show how the location varies as the applied shear-rate is ramped up. At small applied $\dot{\gamma}$, the position of the band fluctuates, although the $\Delta\gamma$ is not large enough for the band to traverse the entire system. As $\dot{\gamma}$ is ramped up, there is a contrast in mobility with the band becoming nearly static at $\dot{\gamma}^*$ where the flow curve has a minimum. This is consistent with our results for the non-affine motion of particles in small systems (see Fig. 21) and can explain the existence of large history effects in the ramping simulations. Only on timescales large enough for the fluid band to traverse the entire system, can structures in the solid band be erased. On smaller time-scales these textures remain and reflect the properties of the system at the previous strain rate probed.

Finally, we explore how the spatial profile of the shear-bands change, when the external strain rate is varied. In the top panel of Figure 24, we show the spatial profile of local shear-rates ($\dot{\gamma}_{\text{local}}$), normalised by the imposed $\dot{\gamma}$. At the smallest shear-rate, we have a very localised band of large fluidization. With increasing shear-rate, we observe that the height of this spatial profile decreases, implying that the fluidized region has less contrast in flow-rate with the solid-like region. Also, the width of this region broadens and the shear-band finally disappears in the regime where repulsion dominates. In all these cases, we have checked and found that the stress generated in the system is spatially uniform, albeit with minor fluctuations.

From the spatial profiles of the shear-bands, we compute the width of the solid-like and fluid-like regions, as well as the interface of the shear-band. This data is shown in the bottom panel of Figure 24. As described above, the width of the solid-like region decreases linearly and, similarly, the liquid-like region increases with increasing $\dot{\gamma}$. On the contrary, the width of the interface remains nearly constant, consistent with earlier numerical observations [41]. Thus it might actually reflect an intrinsic material property, e.g. a shear curvature viscosity [30].

IV. CONCLUSION

We have studied the rheological response of an athermal system of particles, having weak attractive interactions, by scanning across a wide range of packing fraction (ϕ), attraction strength (u) and imposed shear-rates ($\dot{\gamma}$). These extensive simulations reveal that at vanishing shear-rates and weak attractions, a fragile isostatic solid exists even if we go to $\phi \ll \phi_J$. Further, with increasing shear-rates, even at these low ϕ , non-monotonic flow curves occur which (in large enough systems) lead to persistent shear-bands. Our exploration of parameters allows us to draw up the regime in $u - \phi$ where such shearbanding is to be expected, which we observe to be spanning a large parameter window. The non-monotonic flow curves are also associated with a non-monotonic dependence of the macroscopic friction, μ , on imposed shear-rate, with the maximum in the resistance to shear occurring at the exact point where the minimum in the flow curves occur. The low shear-rate regime, where attractive interactions dominate the rheological response, is also characterized by enhanced non-affine dynamics. Finally, we demonstrate that the non-monotonicity in the flow curves is enhanced if one tunes the dissipation timescale to probe the under-damped regime of the dynamics.

The solid-like response of any material is characterized by the yielding stress threshold, σ_y . Similarly, we define a potential energy threshold, E_y . Just like for σ_y , we demonstrate the existence of scaling relationships for E_y . Where $\phi > \phi_J$, $E_y \propto \delta\phi^\beta$, with $\beta \approx 2.1$. Also, for

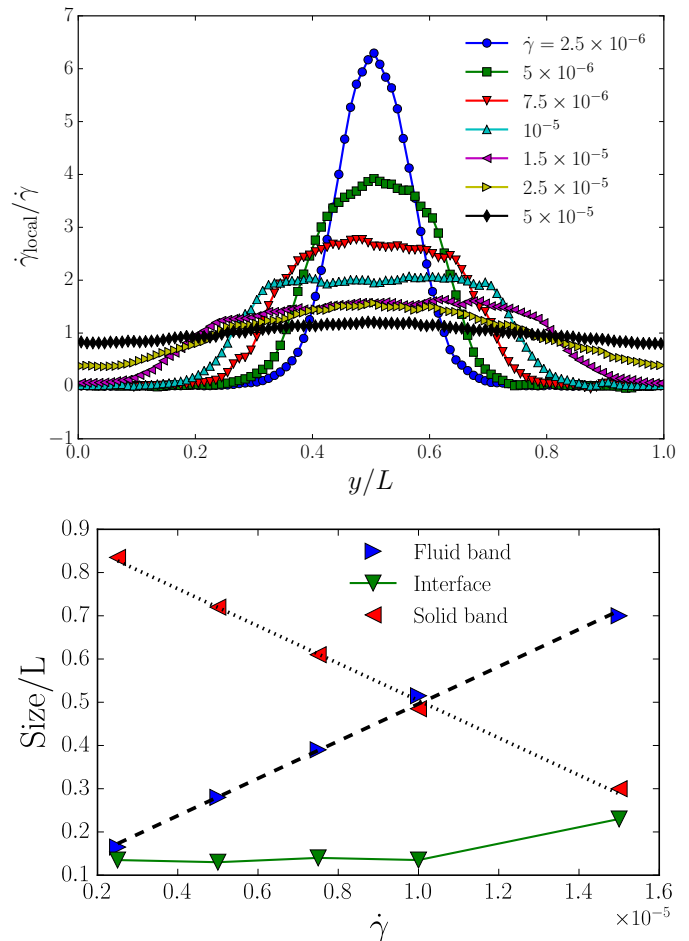


FIG. 24. (Top), centralized shear bands for different strain rates in ramp-up branch. (Bottom), shear bands and the interface width in units of system size, as a function of imposed strain rate $\dot{\gamma}$. The dashed line is the fitted linear function to the size of fluid band, $S_f(\dot{\gamma}) = 4.31 \times 10^4 \dot{\gamma} + 6.51 \times 10^{-2}$. The dotted line presents the fitted function to the size of solid band which is by definition $S_s(\dot{\gamma}) = 1 - S_f(\dot{\gamma})$.

$\phi < \phi_J$, we show that $|E_y| \sim u^2$, i.e. it is determined by the strength of the attractive interaction between the particles.

The macroscopic rheological response of such materials is further probed at the particle level by measuring the structure factor, $S(q)$. It exhibits a peak at small q , implying clustering of particles, at small shear-rates. By studying the partial structure factor, we conclude that the mesoscale clustering induced by shear consists primarily of the smaller particles.

Although non-monotonic flow curves are obtained for $N = 1000$, we need to explore larger system sizes, e.g. $N = 20000$, in order for shear-bands to form. This is due to the fact that sufficiently large systems are necessary for accommodating the spatially heterogeneous flow. Precursors of this banding transition can, however, be observed in a pronounced anisotropy of the particle dynam-

ics, with the deviatoric velocity as well as the diffusion constant in flow direction enhanced as compared to the gradient direction. We also observe that for such large systems, the flow curves deviate from those obtained in smaller systems. Further, similar to recent experiments, hysteretic effects are observed when the applied shear-rate is ramped down and then ramped up. We explain these features with a small mobility of the fluid band leading to prohibitively long time-scales necessary to fully erase any memory present in the solid band. During such a ramping protocol, shear-banding becomes prominent when the strain rate is ramped up, with larger shear-rates necessary to fluidize the solid.

In future, extensive experimental studies are necessary for exploring and validating the observations made via our numerical investigations regarding the rheologi-

cal response of weakly attractive particulate systems. All these studies have focused on athermal suspensions. Further explorations are necessary to check how the flow behaviour changes if one considers Brownian suspensions of such particles, i.e. whether thermal fluctuations modify the response. Studies are also necessary to understand the rheology of such suspensions in the context of more complex flow protocols, which are common in industrial applications.

V. ACKNOWLEDGMENTS

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